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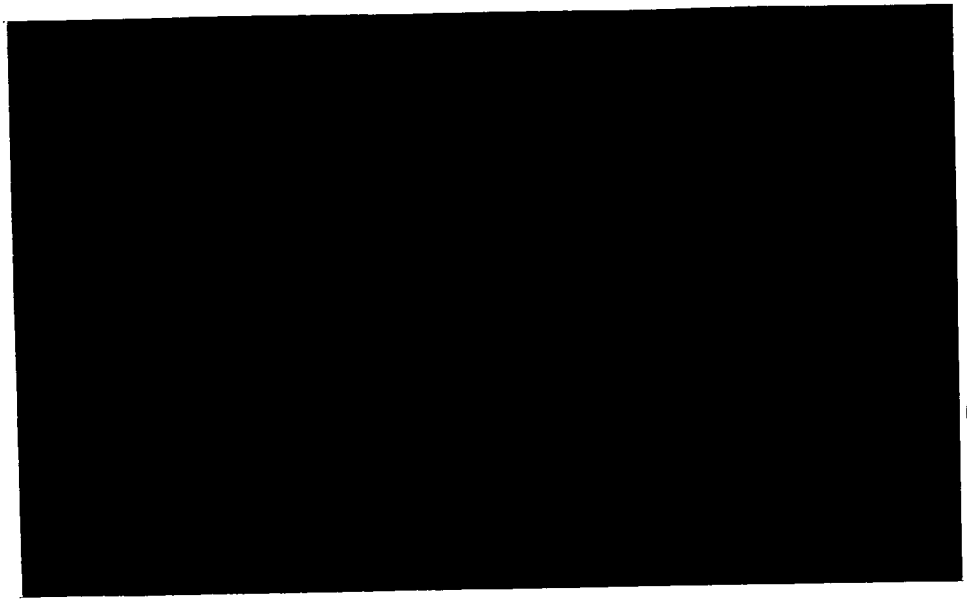

OTS PRICE

XEROX $ 4.60 ph.

MICROFILM $ 1.52 mf.

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ELECTRO-OPTICAL SYSTEMS, INC. Pasadena, California

Quarterly Progress Report
27 September - 27 December 1962

RESEARCH PROGRAM RELATED TO VAPOR THERMIONIC CONVERTERS FOR NUCLEAR APPLICATION

Prepared for

Lewis Research Center
Space Electric Power Office
21000 Brookpark Road
Cleveland 35, Ohio

NASA Contract NAS 3-2529

EOS Report 3410-Q-1

16 January 1963

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1. GENERAL INTRODUCTION

This is the first quarterly report on Contract NAS 3-2529 covering the period from 27 September 1962 through 27 December 1962.

Work on Contract NAS 3-2529 with the Lewis Research Center of NASA was initiated on 27 September 1962. The effort on this contract is being performed in accordance with the contract work statement and EOS technical proposal BD 62-801A, dated 19 June 1962.

The major objective of this research program is to study the effects of long time at temperature on the surface crystal structure of polycrystalline molybdenum substrates and how these changes in surface crystal structure relate to the performance of cesiated molybdenum emitters for use in cesium vapor thermionic converters for nuclear applications. The investigations are primarily limited to molybdenum and/or vapor deposited coatings on molybdenum as a starting point. The decision to limit the investigations to molybdenum substrates was mutually agreed upon after discussions with personnel at Lewis Research Center.

It is known that initially polycrystalline materials which have been processed at temperatures greater than the recrystallization temperature have a preferred surface crystal orientation. The surface of such recrystallized material contains a distribution of a finite number of crystal faces which depend upon: (1) the type of material (for example, its crystal structure), (2) the processing of the material during fabrication (arc cast versus rolling), and (3) the thermal treatment which the material receives after fabrication. It is further known that some body centered cubic materials such as molybdenum and tantalum undergo a process of preferential grain growth as a function of time at temperature beyond the recrystallization point. Many studies have shown the importance of surface crystal structure of a substrate as it relates to the

cesiated emission from that substrate. It is thus clear that it is very important to know how an initially polycrystalline material undergoes surface crystal changes during processing and long term at high temperature in order to quantitatively predict the performance of such substrate materials in practical thermionic converter applications.

The program which we are pursuing in order to gain this quantitative information pertaining to long term effects on the cesiated emission from initially polycrystalline refractory substrates is essentially a 4 point program involving: (1) sample processing investigations, (2) grain growth experiments in vacuum and cesium vapor environments, (3) long term cesiated emission tests from such substrates which are undergoing surface crystal changes and (4) electron emission microscope examinations of initially polycrystalline materials which are operated long times at high temperatures. The main body of the text contains detailed information on the status and results obtained in each of the 4 above mentioned areas.

2. SAMPLE PROCESSING INVESTIGATION

2.1 Introduction

The wide variance of data in the literature pertaining to cesiated emission refractory substrates may, in part, be attributed to the lack of a standard preparation treatment for emission specimens. Additionally, emission data is rarely identified with the crystal structure of the substrate material. It is interesting to note that the emission data gathered by Langmuir and Taylor (Ref. 1) apply only to very special crystal surfaces of tungsten (mainly [110] and [112]). This, of course, is a consequence of the very careful thermal etching at high temperature ($2800^{\circ}K$) which these investigators used to form essentially single crystal tungsten wire. Langmuir and Taylor were very careful to point out the steps involved in properly preparing filaments in order to obtain reproducible cesiated emission data.

The concern about surface crystal structure of so-called "polycrystalline" emitter substrates is best exemplified by an example of considerable significance to practical vapor thermionic converter devices. Pugh and Hibbard (Ref. 2) report a series of experiments on the surface crystal structure of "polycrystalline", recrystallized tantalum sheet which has been rolled from bar stock. They report that the sheet stock developes a surface [112] rolling texture with some [100] and [111] crystal planes in evidence. After annealing, the [112] and [100] crystal orientations disappear, and after recrystallization the [111] crystal planes remain as the dominant orientation. They further state that the entire process of recrystallization in tantalum is probably governed by growth selectivity, wherein one of the crystal orientations in the surface of the material grows at the expense of the other orientations.

In light of the foregoing, one should logically expect substantial differences in the emission density measured on cesiated recrystallized sheet tantalum [111] orientation) and that obtained from wire stock [110] and [112] orientations). Other "rolled" materials recrystallized in different fashions depending on, among other things, the direction of easy slip in the crystal structure. The surface crystal structure of the particular substrate sample to be used for emission measurements must be known before any real scientific sense can be made of emission data obtained from such samples.

One substrate material (molybdenum) is being used in our studies, and the type of molybdenum is limited to the arc cast variety of high purity. We are working with samples which have received two types of processing during manufacture, namely: (1) rolled flat stock which has been heavily reduced (60 percent or more), and (2) round stock which is rolled from arc cast molybdenum rod. All of our flat stock samples are obtained from one sheet of stock and all of our round stock samples are from the same rod.

The chemical cleaning, polishing, and machining of all samples undergoing comparative tests are nearly identical as possible.

The times and temperatures selected for the sample processing and the times and temperatures selected for the remaining studies were arrived at by consideration of the results of at least two studies (Refs. 3 and 4), the partial results of which are shown in Figure 2-1.

2.2 Preparation of Materials

2.2.1 Mill Specifications

As a starting point for material control, we have selected quality mill stock which is low in carbon content and interstitial gas impurities as indicated in Table 2-I. In addition, this stock has received final heat treatment at the mill to achieve an initial controlled grain size in the "as received" condition. Two things are accomplished: (1) a terminal process is specified which minimizes differences in production metal working and (2) a material specification is chosen which is more pertinent to thermionic phenomena than tensile strength, Brinell hardness, etc.

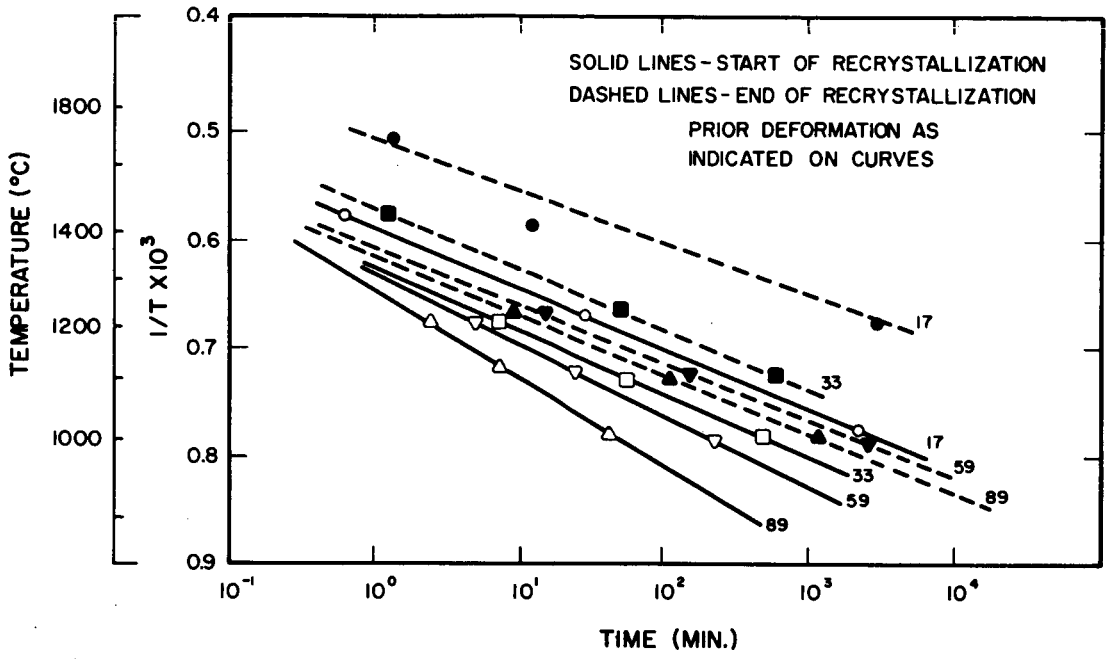

FIG. 2-1 TIME-TEMPERATURE RELATION FOR RECRYSTALLIZATION

TABLE 2-I

Arc-Cast Molybdenum Plate

Mo	99.99 °/o
C	0.006
O_2	0.0006
H_2	0.0001
N_2	0.0002
Fe	0.001
Ni	0.001
Si	0.001
Others	0.0001

Arc-Cast Molybdenum Wrought Bar

Mo	99.9 °/o
C	0.010
O_2	0.0002
H_2	0.0001
N_2	0.0001
Fe	0.001
Ni	0.001
Si	0.001
Others	.0866

On request the mill is supplying an exact history of the received stock from the 12" cast ingot stage to the heavily reduced plate and bar stage.

2.2.2 Machining and Polishing

It has been the experience of the electron tube industry that sulfur-based oils employed in the machining of refractory materials are subsequently difficult to remove. The presence of sulfur compounds in thermionic converters can lead to changes in work function and contamination of the cesium. We accordingly machine molybdenum without any lubricant or with pure mineral oil lubricant. Dry machining offers the best approach to achieve minimum contamination but requires a great deal of time and cost in tooling.

We have observed from experience the adverse effect of certain lapping compounds and polish papers. Silicon and iron bearing lapping compounds, such as once embedded in refractory materials, are exceedingly difficult to remove. The presence of such high vapor pressure materials in the thermionic converter emitter can result in black deposits on the collector during long term operation.

2.2.3 Chemical Cleaning

Starting with select materials and exercising due caution in machining and polishing, we adhere to strict cleaning procedures which have been formulated by close association with the tube industry and allied fields. Appendix A includes the cleaning specification for molybdenum.

2.2.4 Outgassing

Outgassing is normally considered the final step in any preparation of materials for thermionic emission devices. It is part of this program to systematically investigate time at temperature treatment of molybdenum.

2.3 Experimental Procedure

Our samples were prepared in accordance with 2.2 prior to the time at temperature investigations. Temperature measurements were obtained by viewing a properly dimensional hohlraum with a micro-optical pyrometer. Samples were heated by electron bombardment in a vac-ion pumped chamber

which continually operated at 10^{-7} to 10^{-8} mm Hg. with the sample at temperature. Of particular importance is the low hydrocarbon vapor background which is achieved with the use of a vac-ion pumped chamber.

A simple fixture allowed processing temperatures to be attained rapidly. In fact, 1 percent or less of the processing time is involved in the transition from ambient to process temperatures. This is not normally realized in systems which employ induction or resistance heating and more complex jigging. In the latter cases large thermal time constants are encountered which may far exceed the desired processing time.

2.4 Preliminary Results of Processing Investigations

2.4.1 Materials

During the processing of metals into bar or sheet, the material is subjected to severe mechanical working below its recrystallization temperature. This reduction process results in individual crystals within the material being fragmented and broken up into a greatly increased number of smaller crystallites.

As the reduction process proceeds, the internal energy and surface area of the crystallites are greatly increased by the external energy which is added. The uniformity of grain fragmentation during reduction is dependent upon both the degree of reduction, and the cross sectional thickness of the finished article. Rod or bar, for example, as rolled does not exhibit the uniformity of grain across the cross section as does sheet stock of similar dimensions. The reason for this lies in the method of processing. During the sheet rolling operation, the ingot is uniformly deformed across its cross section. On the other hand, during the drawing or rolling of bars, the greatest deformation occurs in the outside fibers, thereby resulting in a non-uniform cross sectional structure. Degree of deformation can generally vary between samples in the same bar.

Figures 2-2a, 2-2b, and 2-3 show the grain structure at the center and outer edge of a molybdenum bar stock sample in the as-received condition. It will be noted that there is little, if any,

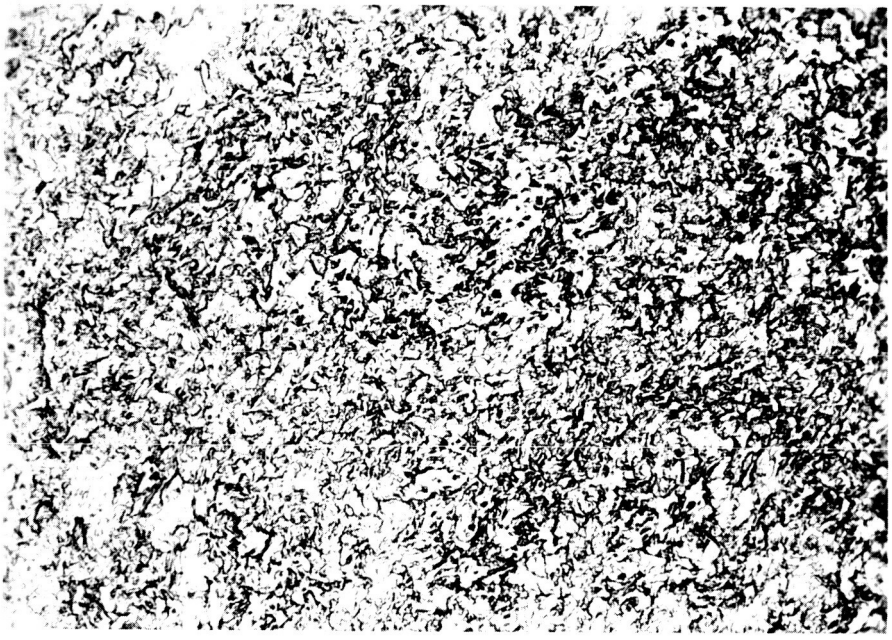

FIG. 2-2a 820-2 MOLYBDENUM BAR - AS RECEIVED - SPEC NO. 257 - MAG 100X OUTER EDGE OF BAR

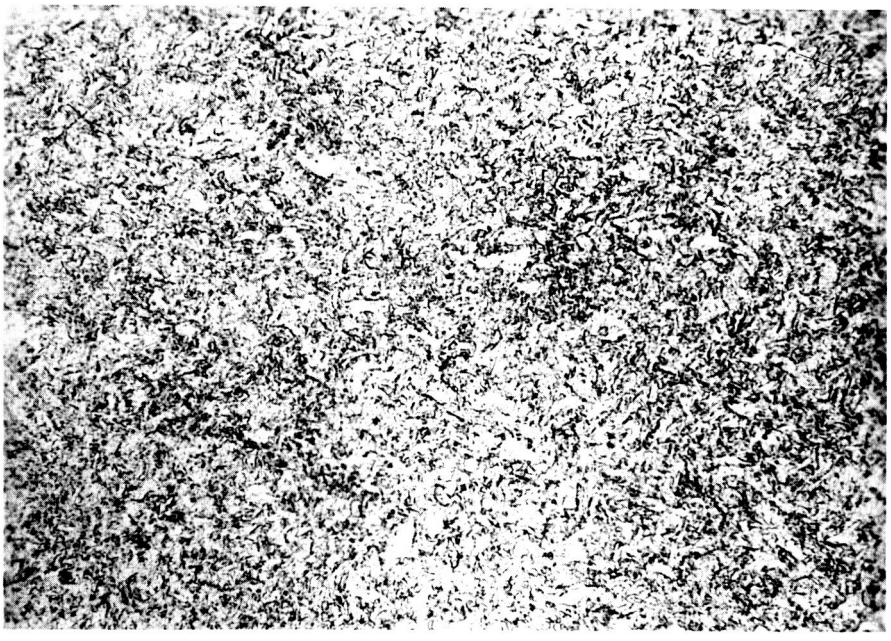

FIG. 2-2b 822-6 MOLYBDENUM BAR - AS RECEIVED - SPEC NO. 257 - MAG 100X - CENTER AREA

FIG. 2-3 759-3 MOLYBDENUM BAR - AS RECEIVED - 600X - BOTTOM FACE - RANDOM SHOT - SAMPLE #2

apparent order. These photographs of unrecrystallized bar stock are in great contrast to those shown in Figure 2-6 and 2-7 for the recrystallized bar stock.

Figures 2-4 and 2-5 show the surface grain structure of the as-received plate stock. This material has received a one hour, 2200°F, vacuum anneal prior to delivery. The surface grain size is approximately A.S.T.M. No. 5.5 and the material has obviously been completely recrystallized by the annealing treatment. The pictures are from two different plates and indicate the much greater material uniformity which is achievable in flat stock as opposed to bar stock.

2.4.2 Recrystallized Materials After Initial Processing

When cold worked material is heated above a certain minimum temperature, recrystallization of the material is initiated about many centers of nucleation, with the result that grain growth occurs. As recrystallization proceeds certain of the crystals grow at a more rapid rate than those adjoining with the result that some of the smaller crystals become "absorbed" by the larger ones. If the recrystallization process is carried out sufficiently, the grain eventually reaches a size beyond which the growth rate becomes extremely slow. This increase in grain size results primarily from the material seeking a minimum energy state. One of the objectives of this program is to study the possibility of achieving a terminal grain growth in molybdenum during processing.

After the proper polishing procedures, the surface of the metal sample is etched with various chemical reagents to render the grain structure visible under the microscope. This etching procedure makes visible the grain structure as a result of the differential metal solution rate existing on different crystallographic planes. Note the three distinct shades of grey, for example, on the etched samples in Figures 2-6 and 2-7. One of the major objectives of this investigation is to determine the change in surface crystal structure of a recrystallized molybdenum substrate with time at temperature. The etch shade pattern clearly brings out the change in structure. For example, one expects a

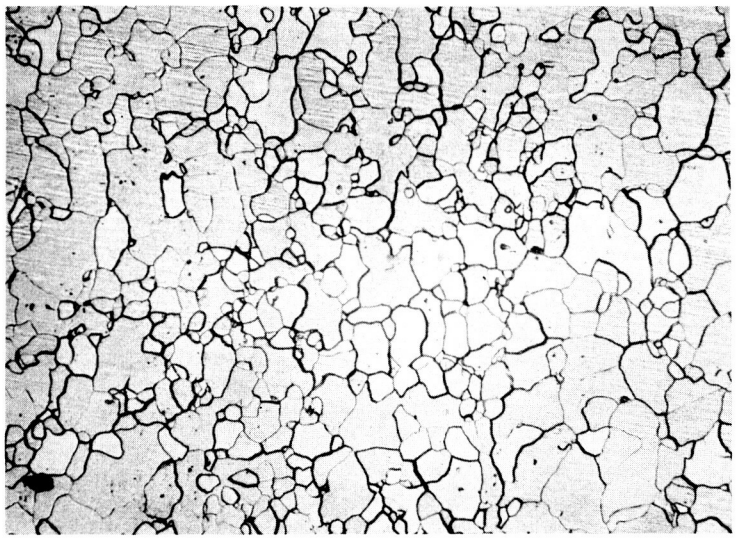

FIG. 2-4 C7092-4 MOLYBDENUM FLAT STOCK SAMPLE (AS RECEIVED) ONE HOUR VACUUM ANNEAL AT 2200°F (PLATE 4) MAG 100X

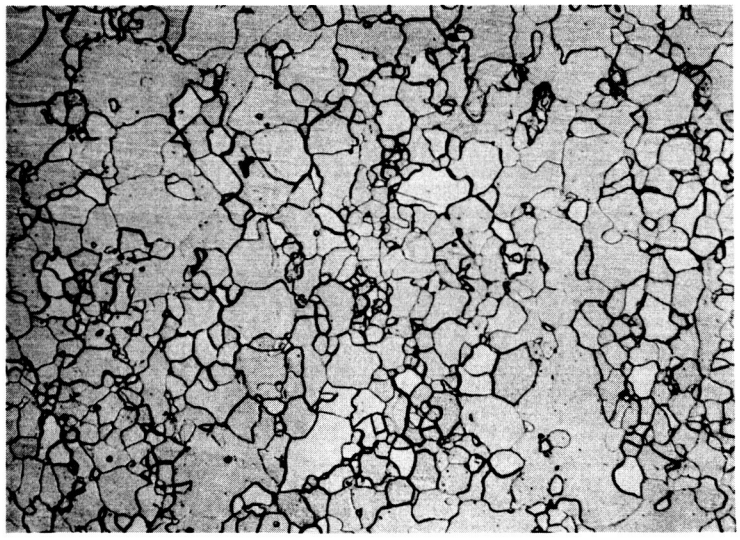

FIG. 2-5 C7092-2 MOLYBDENUM FLAT STOCK SAMPLE (AS RECEIVED) ONE HOUR VACUUM ANNEAL AT 2200°F (PLATE #2) MAG 100X

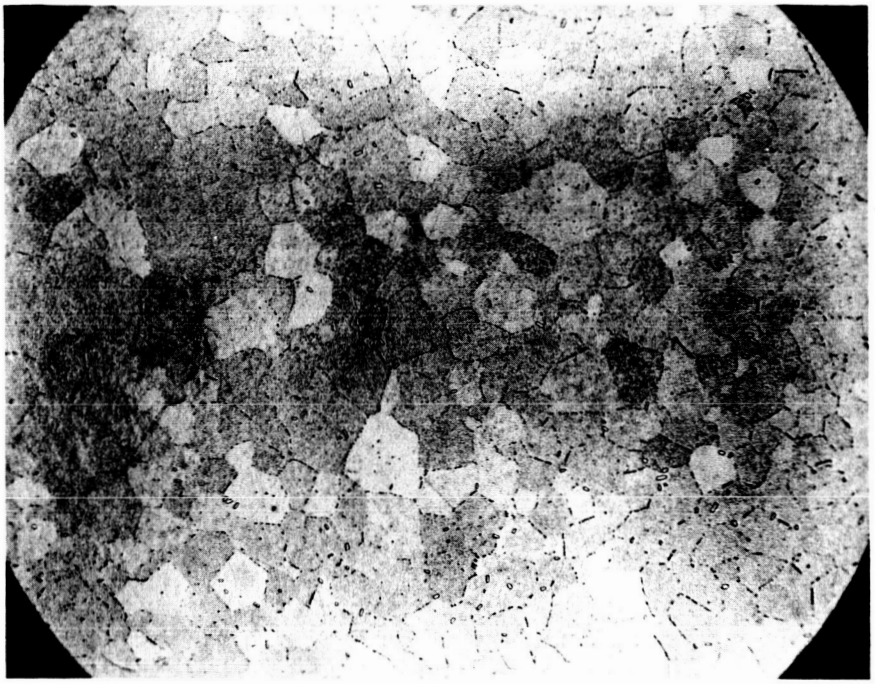

FIG. 2-6 785-3 MOLYBDENUM SAMPLE # 1 - AFTER PROCESSING 10 MINUTES AT 1700°C VIEW - CENTER OF SAMPLE MAGNIFICATION 60X

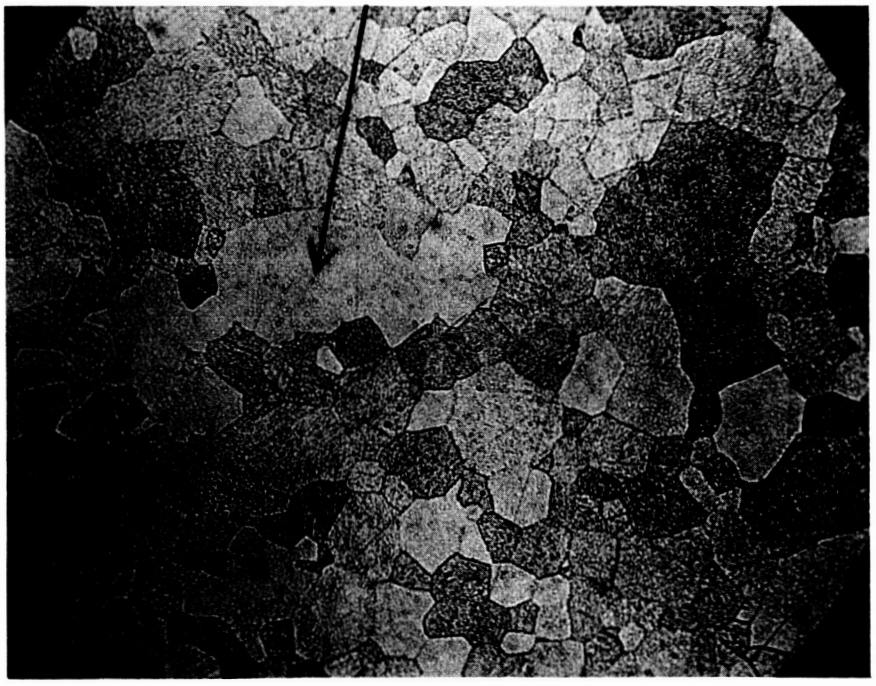

FIG. 2-7 789-7 #2 MOLYBDENUM SAMPLE HEATED FOR TWENTY MINUTES AT 1700°C VIEW - CENTER OF SAMPLE MAGNIFICATION - 60X

different etch rate for each crystal face and hence could interpret the picture of Figure 2-7 to mean that only three crystal faces are dominant in the surface of recrystallized molybdenum bar stock. Comparison of Figure 2-7 with Figure 3-1 and Figure 3-2 which is the same material after operation at temperature for 100 hours, indicates that not only has further grain growth taken place with the longer term operation but also that now there are only two etch shades present. This indicates qualitatively that preferential grain growth has occurred and one surface crystal orientation has disappeared.

Figure 2-7 also indicates the presence of one very large elongated grain (see arrow) which appears to have grown by the process of absorbing smaller adjacent grains of the same orientation.

As recrystallization and grain growth occur within the metal, impurities such as carbides tend to be rejected at the grain boundaries. If the temperature is sufficiently high, and the heating time extended, the carbides become spheroidized into a "bead-like" structure surrounding each grain. This effect can be seen in Figure 2-6.

The presence of small quantities of impurities such as carbon, nitrogen, silicon, etc. within the molybdenum will result in the presence of a considerably greater quantity of the refractory compound. For example, the compund Mo_2C contains 6 percent C. Therefore, the weight percentage of Mo_2C formed will exceed that of carbon by approximately 16 times.

3. GRAIN GROWTH INVESTIGATION

3.1 Introduction

We are investigating the grain growth in bar and plate stock molybdenum which occurs as the result of long term operation at high temperature. The investigation is being conducted in vacuum and cesium vapor environment as a check on the possibility that the presence of cesium vapor might effect surface structure during long times at high temperature.

3.2 Vacuum Environment Results

Two molybdenum bar stock samples were investigated for grain growth at 1800°K for operating times of 100 hours. The investigation was conducted principally to compare the grain size of these samples with samples processed at higher temperature but shorter times. Figures 3-1 and 3-2 are representative of two grain growth specimens which operated for 100 hours at 1700°K and 1800°K respectively. Temperature measurement techniques and vacuum environment conditions were described in Section 2.3. The average grain size of these samples varies from .25 mm to .30 mm as compared to .05 to .08 mm for process samples operated at 1973°K for 1/2 hour. It is interesting to note that in a properly light-relieved photograph such as Figure 3-1 there are fewer shades of gray than in the process photographs. This indicates the tendency of recrystallized materials to abandon their once polycrystalline state in favor of an essentially single crystal type of surface structure.

In addition to the grain growth, we have observed a significant evaporation of material from molybdenum samples during time at temperature (1800°K) studies over a 100 hour period. These coatings have been analyzed to establish the constituent elements and are reported in Table 3-I. One expects that the trace impurities of low melting point will constitute a greater percentage of the evaporated coating than they did of the emitter

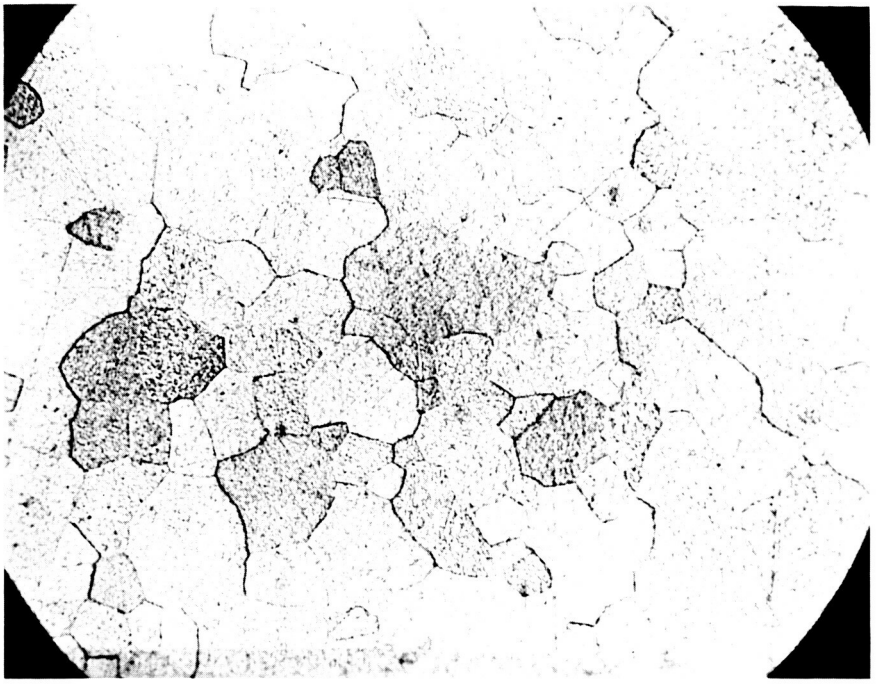

FIG. 3-1 763-7 MOLYBDENUM BAR 102 HOURS AT 1800°K MAGNIFICATION 60X

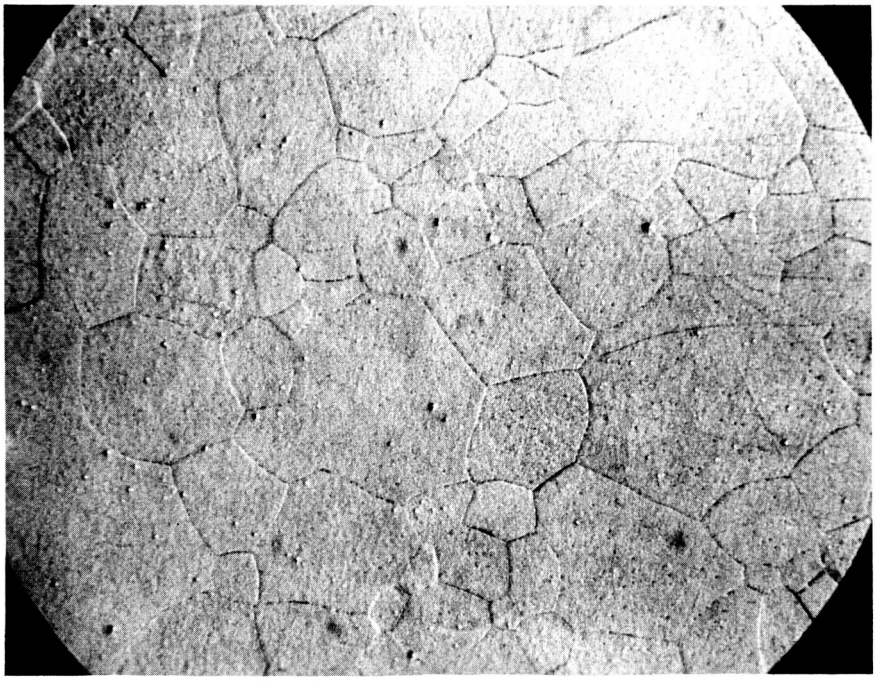

FIG. 3-2 839-7 MOLYBDENUM BAR 102 HOURS 1900°K MAGNIFICATION 60X

TABLE 3-I

EVAPORATED COATING ANALYSIS

Molybdenum Emitter Sample

(100 hour test at 1800°K)

Mo	99.76 °/o
Ag	.084 °/o
Si	.079 °/o
W	.053 °/o
Cu	.026 °/o
Fe	Nil
Mn	Nil
Ta	Nil
Cd	Nil

sample. It is seen from the analysis that trace elements having a high vapor pressure (low melting point) have been differentially outgassed from the sample. Within the limits of measurement accuracy the evaporated coating is 99.8 percent molybdenum.

Weight gain measurements on the collecting surface have established order of magnitude agreement with calculated values based upon the vapor pressure of molybdenum. An extensive investigation of evaporation weight loss is not presently part of our program.

It is our contention that the evaporation of as little as a 200 Å layer of molybdenum from the emitter in the presence of cesium vapor could result in an opaque coating of high emissivity being formed on the collector of a practical thermionic converter. Such a collector deposit could reduce device efficiency by 20 or 30 percent in certain instances. We feel that weight loss data for thermionic converter emitter materials is more meaningful when translated into angstrom layers/cm^2/1000 hours, and then interpreted in terms of its significance to long-time device efficiency. We have demonstrated experimentally that coating layers approximately five times this acceptable thickness (i.e., 1000 Å) are evaporated from an 1800°K molybdenum emitter in times as short as 100 hours. These results indicate an absolute maximum useful life of 100 hours at 1800°K for a molybdenum emitter if peak thermionic converter performance is a consideration.

These results and maximum allowable operating temperatures for molybdenum are in marked contrast to lifetime calculations which have appeared in the literature. In many instances, investigators have arbitrarily established .001 inch/cm^2/yr. loss of material as an acceptable level which would not seriously change dimensions, spacings, or mechanical strength in a thermionic converter structure. The .001 inch/cm^2/ yr. evaporation corresponds to a layer approximately 2.5×10^5 Å thick. Since it is known that 200 Å layer coatings evaporated in the presence of high pressure vapor produce an opaque coating of high emissivity, it would appear that a .001 inch/cm^2/yr. criterion is a

factor of 1000 times too large for practical use when the physics of the converter is used as a criterion. Viewed differently, an emitter material that was evaporating material at the rate of .001 inch/cm^2/yr. could conceivably decrease the converter efficiency by 30 percent in 10 to 100 hours of operation.

3.3 Cesium Environment Vehicle

The design of a cesium environment container and the attendant furnace has been completed. The experimental vehicle, as shown in assembly drawing (Figure 3-3), will be fabricated of outgassed, high purity tantalum sheet and tubing. As indicated, a section of the container is reduced in wall thickness. The smaller wall cross-section impedes the heat conduction from the furnace region allowing a cool portion of the vehicle to condense evaporation products. This is necessary to preclude the continued interaction of impurities with the samples. A section of the container will operate between 1100°C and 1400°C, a temperature at which tantalum efficiency getters oxygen and nitrogen if present. This will further guarantee a high degree of sample cleanliness.

The fabrication of the cesium environment test vehicle is in progress. Machined parts are presently being joined in a high-quality dry box by inert arc welding. Resultant welds are free of oxide and nitride formations.

After construction, the container will be sealed off at a pressure of less than 10^{-7} mm Hg. in the absence of any hydrocarbon vapors. 99.9 percent cesium will be vacuum distilled into the tantalum container to furnish the cesium environment. Glass ampoules will be avoided because of the known cesium vapor interactions with all types of glass at high temperature (greater than 200°C).

A cesium reservoir appendage will serve to control cesium pressure during operation. The appendage will be operated at temperatures which will insure cesium pressures equivalent to those desired in the emission test vehicle (i.e., in the millimeter range).

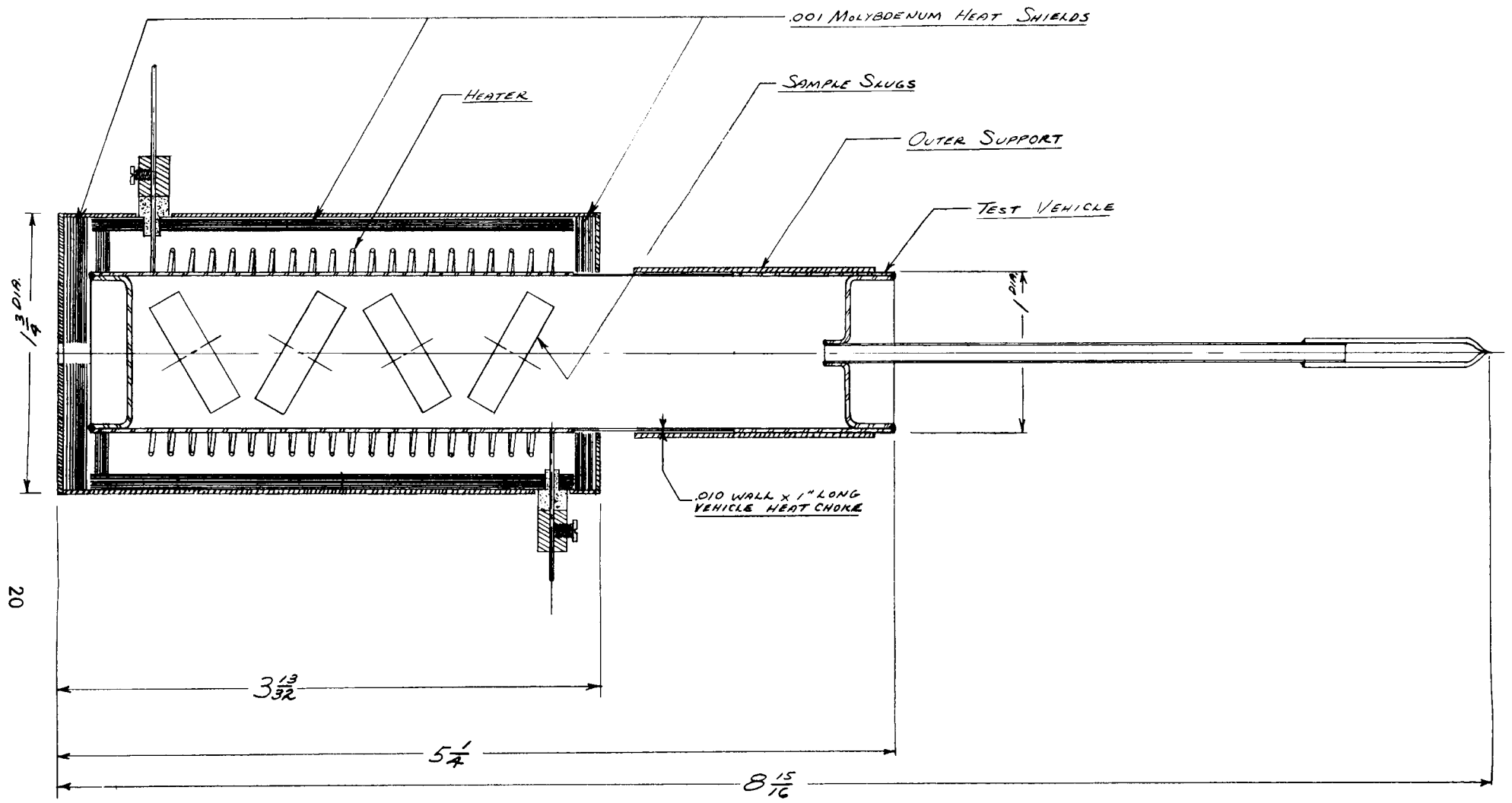

FIG. 3-3 CESIUM ENVIRONMENT TEST VEHICLE FOR GRAIN GROWTH INVESTIGATION

REQ'D	ITEM	PART NO.	DESCRIPTION	SIZE	MATERIAL	SPECIFICATION

LIST OF MATERIALS

UNLESS OTHERWISE NOTED:
1. LINEAR DIMENSIONS IN INCHES.
2. TOLERANCES:

DECIMAL	FRACTIONS	ANGULAR
± .010	± 1/16	± 1/2°

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VEHICLE FOR GRAIN ...

SECURITY CLASSIFICATION	SIZE	NUMBER	REV.
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4. CESIATED EMISSION INVESTIGATION

The test vehicle for performing emission measurements on cesiated molybdenum substrates has been designed and is shown in Figure 4-1. The range of pertinent parameters over which the vehicle will operate is listed below:

1. $700^{\circ}K < T_{emitter} < 1900^{\circ}K$

2. $470^{\circ}K < T_{cesium} < 630^{\circ}K$

3. $10^{19} \frac{atoms}{cm^3/sec} < \text{Arrival Rates} < 10^{21} \frac{atoms}{cm^3/sec}$

4. 10^{-2} amps/cm^2 $< j_{emitter} <$ 20 amps/cm^2

With proper potentials applied to the guard ring it should be possible to measure the emission current from just the top surface of the emitter sample. The area of the face of the sample will be approximately 2.5 cm^2. The sample will consist of emitters fabricated from (1) molybdenum flat stock which has been shown (Ref. 5) to have predominately $[100]$ and $[100]$ crystal orientations in the recrystallized sample surfaces, and (2) from molybdenum rod stock which has been shown to have predominately $[100]$ crystal surfaces in the rod cross section surface. There is evidence in the literature that some materials have exhibited almost an order of magnitude difference in electron emission at the same cesium arrival rate from various preferred crystal faces (Ref. 6).

Some of the more important design considerations of electrode spacing, emitter lead, and cesium reservoir, are discussed next.

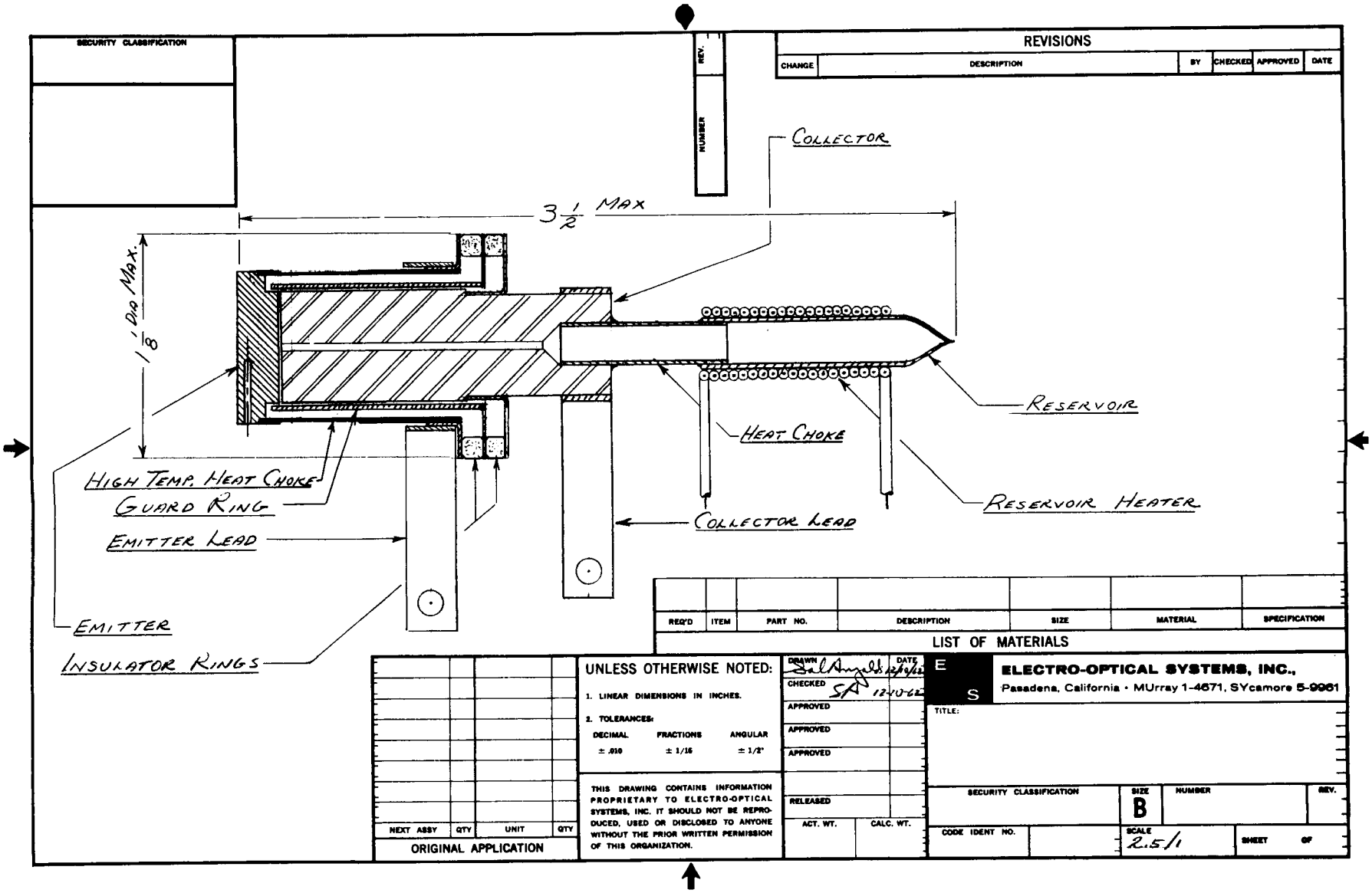


FIG. 4-1 EMISSION TEST VEHICLE

4.1 Electrode Spacing

We have chosen an emitter-collector spacing of .010 inches based on experimental evidence from cesium discharge devices (Ref. 7). The guard ring-collector distance and guard ring-emitter distances have also been assigned a spacing of .010 inches to eliminate the possibility of spurious emission being measured from different "pd" paths in the vehicle. In this manner we should be able to accurately define an emitting area to which the measured current can be correctly ascribed. The definition of an accurate emission area is very important since it is the current density which most accurately describes the emission capability of a particular emitter.

4.2 Emitter Lead

The emitter lead of this device should satisfy these basic requisites:

a. Minimize heat flow from the emitter to seal region.

b. Conduct large values of current (30 to 40 amps) without excessive I-R drop.

Unfortunately, these requirements are in opposition since any conceivable scheme of reducing the heat flow will tend to increase the electrical resistance.

We are not, however, basically concerned with device efficiency in this study and, therefore, moderate heat losses can be sustained from the emitter in the interest of facilitating emission measurements. Measurements have been taken on the emitter lead which indicate a 30 millivolt drop from 50 amperes of current flow. For the vehicle design this is equivalent to 15 watts of thermal loss.

4.3 Reservoir

The .010 inch spacing between electrodes is much greater than the atom-atom mean free path for cesium at pressures of the order of 1 mm Hg. The high collision frequency between atoms thus establishes a constant pressure throughout the device and corrections to the cesium arrival rate are necessary for the emitter surface temperature. We will interpret our emission measurements in light of this correction.

Since the cesium reservoir temperature should accurately govern the device pressure, it must be designed to operate independently of other areas in the device. We will, therefore, operate the vehicle at a substantially different temperature than the cesium reservoir by separating the two with a heat choke.

All materials have been ordered in the support of the processing, fabrication and assembly of this vehicle. The essential steps in the fabrication and assembly are reviewed briefly.

a. Processing and Cleaning

All device structural materials, including the emitter, are certified "Electron Tube Grade" to insure minimum impurity levels. With tube grade materials as our starting point, we adhere to strict cleaning procedures which have been formulated by close association with the tube industry and allied fields. After mechanical and chemical cleaning all materials are outgassed at temperatures well above their normal operating temperature in the device. For example, the tantalum guard ring while operating at collector temperatures of 800 to 1000 ^{o}C would be outgassed at 2000^{o}C or higher in a vacuum environment of 10^{-6} mm Hg, since it is known that trace metallic impurities and oxygen are not efficiently removed at lower outgassing temperatures.

b. Metal-Ceramic Sealing

A critical step in a cesiated emission test vehicle assembly is the metal to ceramic seal that provides electric isolation of the emitter, collector, and guard ring. This seal must be vacuum tight and resistant to the corrosive effects of cesium vapor in the pressure range of interest. The thermal expansion characteristics of both ceramic and structural materials must be matched in the temperature range of operation to minimize seal stress. In general, commercially available seals are not able to satisfy these criteria. We will perform the ceramic sealing operation in house for the fabrication of the emission test vehicle. To this end we have completed test sealing operations in support of the device fabrication.

c. Brazing and Welding

To insure a reliable joint between the tantalum envelope and molybdenum emitter, we will either electron beam weld the assembly or heliarc weld the parts in an "ultra-clean" dry box. Preliminary inert-arc welds of refractory metals pertinent to the design have been successful.

Where brazing operations are required, we will employ high temperature, low vapor pressure braze materials with the braze made in a vacuum environment. No flux will be used, nor will any type of oxidizing atmosphere enter into the brazing operations.

5. ELECTRON MICROSCOPE

5.1 Introduction

The electrons emitted thermally from a sample at high temperature can be used to provide a magnified image of the surface by accelerating them through an electrostatic lens and focusing them on a fluorescent screen. Since the emission is dependent upon (1) the crystal orientation of the exposed crystal faces, (3) the surface texture, and (3) the presence of impurities on the surface, the image formed by the electrons will exhibit these important features. Observations can, therefore, be made on surface structure and its time and temperature variations. Accordingly we have designed an emission microscope to supplement the investigations of surface crystal changes during long-time operation at high temperature.

Historically, one of the first electron microscopes consisted of a hot cathode and an electrostatic lens to form a magnified image of the cathode surface on a phosphor screen. The electrostatic lens in the present program is of the type described by Johannson (Ref. 8) and was selected because of its simplicity of design, ease of outgassing and the fact that there is some available information in the literature with which to guide our design.

Effort this first quarter has been directed toward selecting a lens configuration, designing a system of support for the lens, and a means of moving the emitter relative to the lens. Figure 5-1 shows the electron emission microscope assembly drawing.

5.2 Emitter Assembly

The purpose of the three axis movable emitter is to allow focusing of the lens as well as scanning of the emitter surface. It is anticipated that emitter grain sizes from 0.8 mm up to a centimeter will be obtained, the latter being beyond the field of view of the lens.

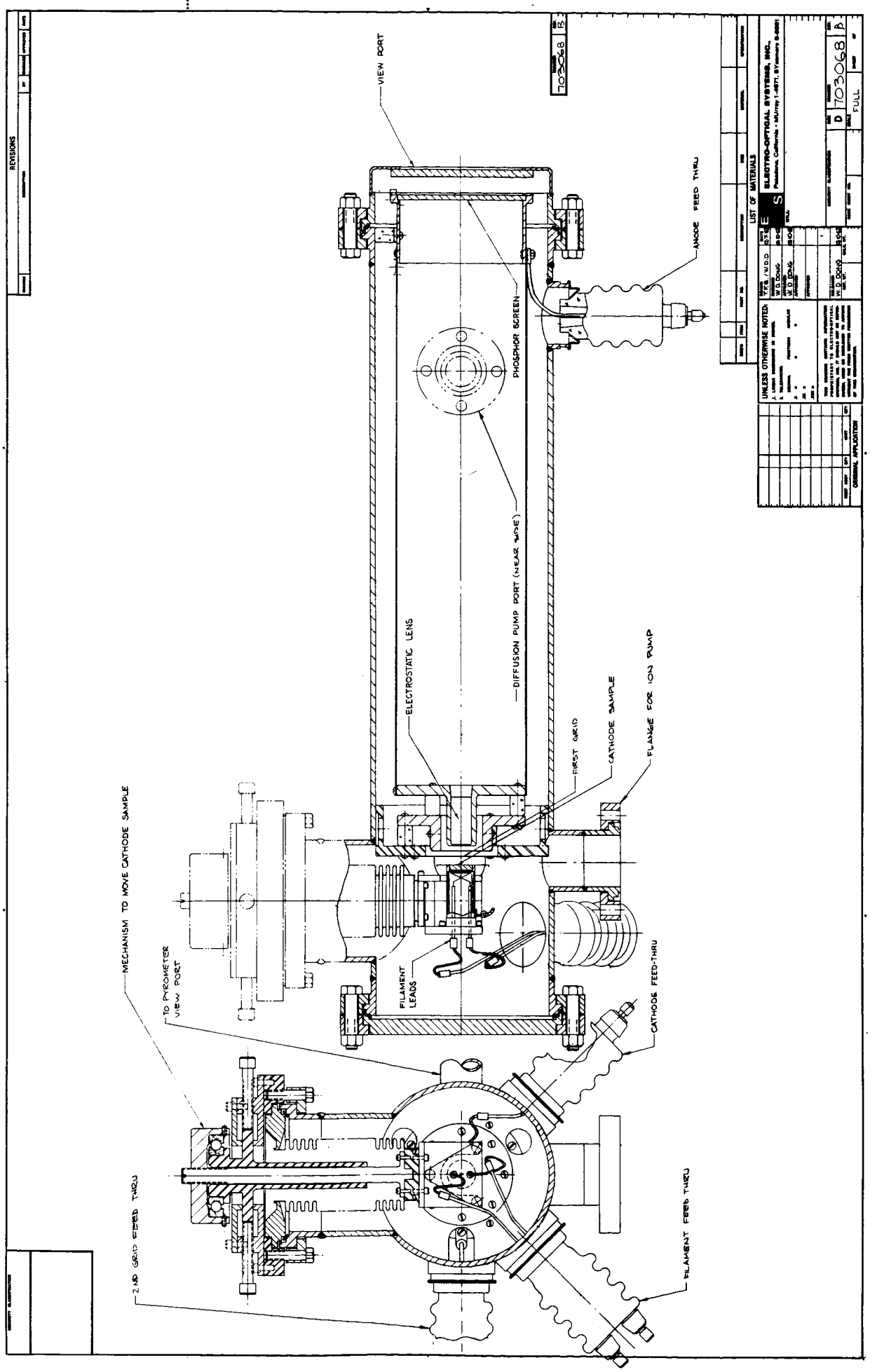


FIG. 5-1 ELECTRON EMISSION MICROSCOPE ASSEMBLY

Figure 5-2 shows a preliminary design of an emitter and lens system mounted in a test microscope set-up which was operated in a glass bell jar system. Details of construction of the emitter assembly are shown in Figure 5-3. The emitter was readily brought to a temperature of 1400°C with the application of approximately 200 watts of bombardment power. This design will be used in the final emitter structure.

5.3 Electrostatic Lens

The test set-up of Figure 5-2 also provided an opportunity to check the heating of the lens electrodes. Heating of the lens electrodes brought about by radiation and electron bombardment could give rise to buckling and spacing changes. Figure 5-2 shows the first lens configuration. This particular lens configuration remained at a temperature less than 700°C when the emitter was operated at a temperature of 1400°C. The spacing between emitter and first grid was 0.1 inch and the accelerating voltage was one kilovolt. No visual evidence of lens buckling was observed under these conditions. However, considerable difficulty was encountered in aligning and preserving the spacings, which suggests that a more rigid design is required. In the modified design a lower operating temperature is achieved by heat sinking which reduces thermal expansion in the mounting. The preliminary design used flexible tabs to take up the thermal expansion differences.

5.4 Phosphor Screen

A simple method of applying the phosophor to the glass window is being explored. The method consists of applying the phosphor in a binder directly on a glass plate. For the experimental set-up, the glass plate with phosphor coating was heated with an infrared lamp to dry the binder thoroughly before placing the screen in the microscope. The phosphor and lens system is currently undergoing test in the bell jar system.

If the binder technique of applying the phosphor proves to be unsuccessful, the more common method of applying the phosphor to the

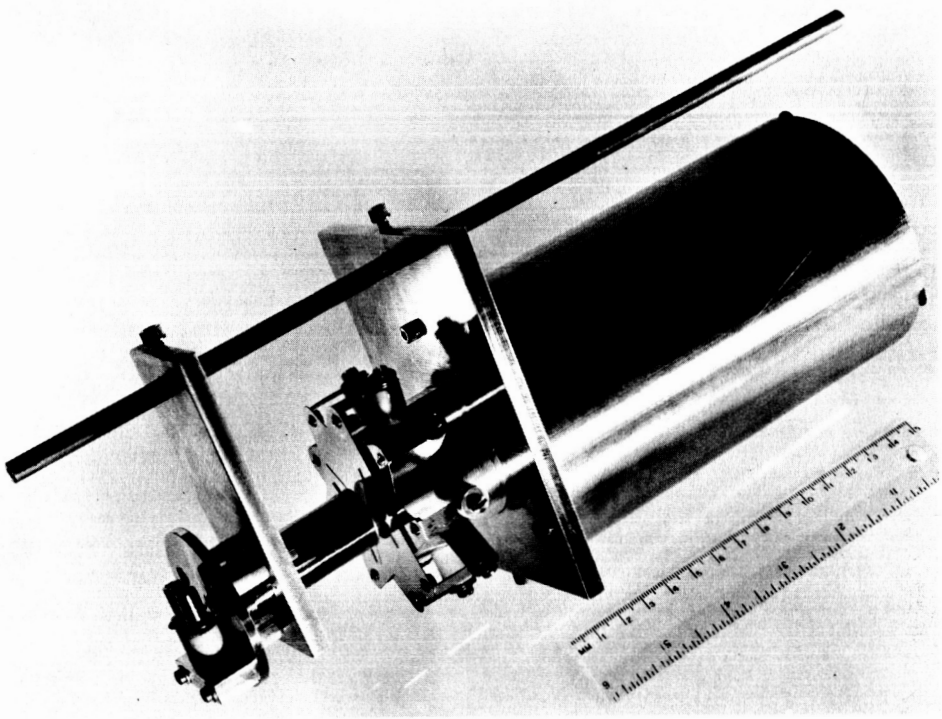
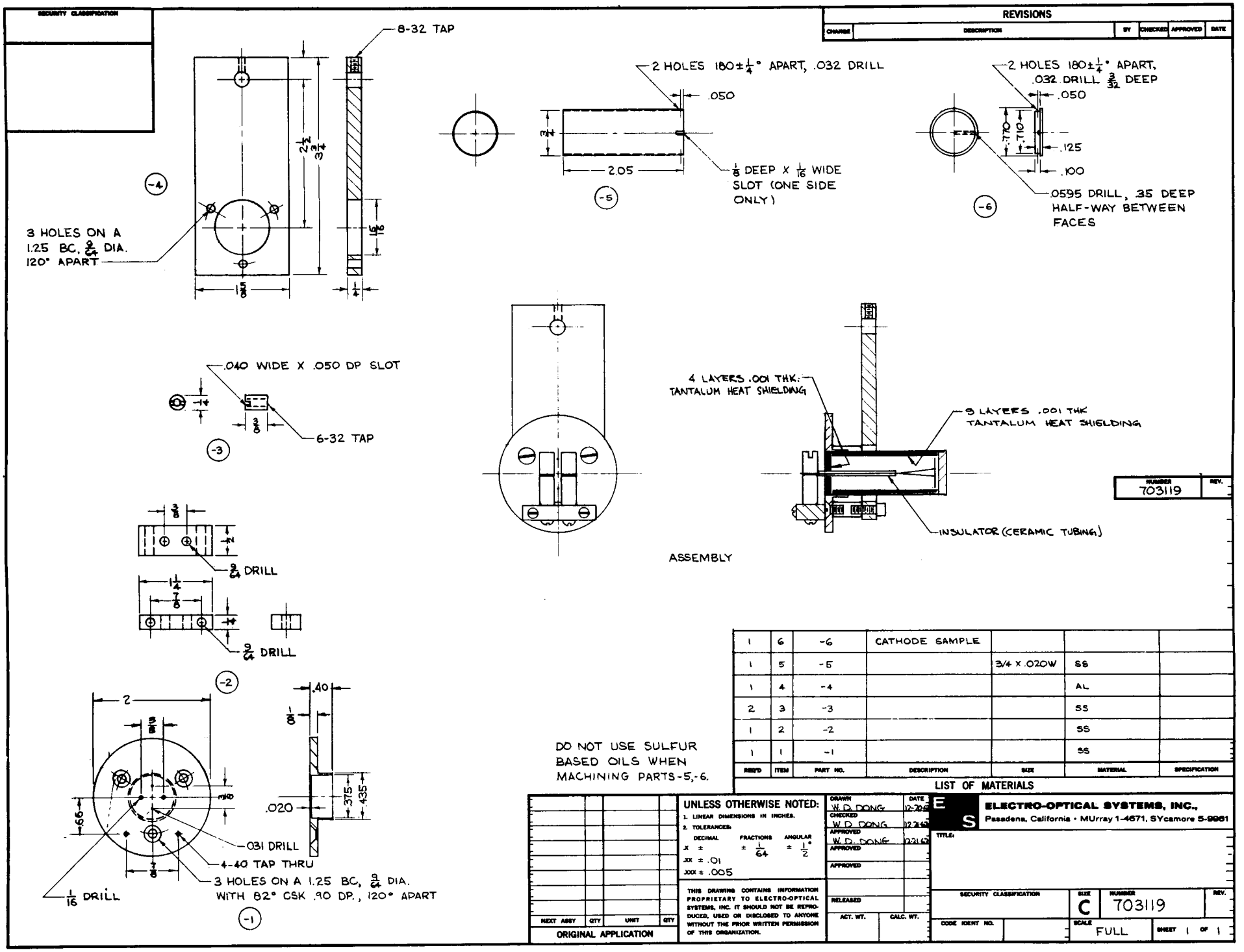


FIG. 5-2 ELECTRON EMISSION MICROSCOPE TEST SET-UP



REQ'D	ITEM	PART NO.	DESCRIPTION	SIZE	MATERIAL	SPECIFICATION
1	6	-6	CATHODE SAMPLE			
1	5	-5		3/4 X .020W	SS	
1	4	-4			AL	
2	3	-3			SS	
1	2	-2			SS	
1	1	-1			SS	

LIST OF MATERIALS

UNLESS OTHERWISE NOTED:
1. LINEAR DIMENSIONS IN INCHES.
2. TOLERANCES:
DECIMAL: .X ±, .XX ± .01, .XXX ± .005
FRACTIONS ± 1/64
ANGULAR ± 1/2°

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	W. D. DONG	W. D. DONG	W. D. DONG			
DATE	12-20-62	12-21-62	12-21-62			

ACT. WT. | CALC. WT.

NEXT ASSY | QTY | UNIT | QTY

ORIGINAL APPLICATION

ELECTRO-OPTICAL SYSTEMS, INC.
Pasadena, California • MUrray 1-4671, SYcamore 5-9961

TITLE:

SECURITY CLASSIFICATION | SIZE C | NUMBER 703119 | REV.

CODE IDENT NO. | SCALE FULL | SHEET 1 OF 1

FIG. 5-3 CATHODE SAMPLE HOLDER (TEST SET-UP)

glass by settling will be employed. If accelerating voltages in excess of 10 KV are needed to excite the phosphor, the screen will be aluminized in order to reduce secondary emission effects and to enhance the image.

5.5 Vacuum Chamber

The vacuum chamber which houses the bakeable microscope is of stainless steel construction and is presently in the process of fabrication. The entire microscope system will be baked out at 400°C while it is connected to a cold trapped diffusion pump system in order to remove occluded gas from the walls. The microscope will be pinched off from the diffusion pump and vacuum will then be maintained by a vac-ion pump during operation of the emitter. Ultra-high vacuum techniques are thus used to obtain the cleanest possible system during the time at temperature investigations.

5.6 Power Supplies

Emitter heating power will be provided by a 16 ampere, 10 volt filament supply and a 200 milliampere, 1 kilovolt electron bombardment supply. The supply is being fabricated.

The electrostatic lens supply is fairly simple in theory. Essentially it consists of a 10 kilovolt supply, a 15 kilovolt supply, and a voltage divider. Its complexity in a practical sense is due to the precautions which must be taken to prevent voltage breakdown of the various components. The lens supply is in the process of being fabricated.

5.7 Summary

5.7.1 Electron Emission Microscope Imaging System

The preliminary lens system, emitter sample holder, and phosphor screen for the electron emission microscope have been assembled and are in the process of being checked out in a bell jar system.

5.7.2 Electron Emission Microscope Device

The U.H.V. chamber for housing the emission microscope has been designed and is in the process of being fabricated.

5.7.3 Test Apparatus

The auxiliary emitter heating and lens accelerating power supplies have been designed and are being fabricated.

5.7.4 Materials

All critical materials necessary for the fabrication of the electron emission microscope have been acquired.

6. ANALYSIS, CORRELATION OF DATA, AND SUMMARY

This program of research pertinent to thermionic converters for nuclear application is proceeding according to schedule. Accordingly, no great mass of data will be available for analysis and correlation until late in the next quarter. A brief summary of the significant experimental results achieved to date follows.

6.1 Demonstration of Grain Growth During Time at Temperature Tests

We have demonstrated that significant grain growth occurs during 100 hour operation of molybdenum emitter samples at temperatures as low as 1800°K. The samples which were obtained from arc cast molybdenum bar stock had been outgassed at 1973°K for 30 minutes before the grain growth experiment began.

6.2 Demonstration of Change in Surface Crystal Structure During Time at Temperature Tests

Etch patterns have been observed on polycrystalline molybdenum emitter samples after recrystallization and after 100 hour operation at 1800°K which definitely indicate a selective grain growth characteristic in the surface of arc cast molybdenum bar stock. One expects different crystal faces to chemically etch at different rates, and thus the change in the number of etch shades on a specimen during long term tests at high temperature (1800°K) is a direct qualitative indication of the change in the number of crystal faces present in the sample surface.

6.3 Demonstration of Evaporation Rate of Molybdenum

We have obtained preliminary data which indicate that the evaporation rate of molybdenum at 1800°K is such that it may be sufficient to prohibit the achievement of highest possible efficiency and power output from a cesium vapor thermionic converter utilizing a molybdenum emitter.

6.4 Demonstration of the Preliminary Feasibility of Electron Emission Microscope Design

Preliminary tests of the electron emission microscope design in a bell jar system have been made. The results indicate that our general design approach to the microscope is adequate.

7. PROGRAM FOR THE NEXT INTERVAL

7.1 Processing Investigations

The sample processing investigations will be completed during this quarter. We feel that we will have arrived at a schedule for time and temperature processing of the molybdenum emitter samples which should allow two objectives to be achieved. The first of these objectives is the achievement of complete outgassing of the substrate sample during the outgassing run. The second objective is that of achieving a time at temperature outgassing schedule which accomplishes outgassing without seriously affecting the surface of the sample. One should not outgas molybdenum, for example, at temperatures much beyond 2100°K in order to limit the gross evaporation of molybdenum from the surface of the sample. Outgassing temperatures on the order of 2300°K may seriously change the gross properties of the surface of the sample due to thermal etching and differential evaporation from various crystal faces in the surface. It is important to note, however, that as the studies progress and we acquire more definitive information on the surface properties of substrates, we may be able to deliberately grow into the surface desired crystal faces by the process of thermal etching during the initial outgassing steps in the materials processing schedule.

7.2 Grain Growth Experiments

The fabrication of test vehicles for the grain growth experiments will be completed during this quarter. Time at temperature experiments pertaining to the grain growth in molybdenum substrates in both vacuum and cesium vapor environments will begin during this quarter. As samples of substrates which have undergone grain growth become available, the replica studies of the surface will begin. It is expected that the replica studies will begin during the latter part of the quarter.

7.3 Emission Tests

The fabrication of a cesiated molybdenum emission test vehicle will be completed during the coming quarter together with the associated apparatus involved in obtaining cesiated molybdenum emission data at high cesium arrival rates. It is expected that cesiated emission experiments will begin during the latter part of this quarter.

7.4 Electron Emission Microscope Experiments

During the coming quarter tests on a bell jar mock up of the aperature system for the electron emission microscope will be completed. The data forthcoming from the bell jar tests of the lens system will be used to make final design modifications of the electron emission microscope which will be fabricated during this quarter. It is expected that vacuum emission tests on prepared substrates will begin during the latter part of the quarter.

7.5 Analysis

As significant quantities of data become available during the latter part of this quarter it is anticipated that a considerable amount of time will be spent in the analysis and correlation of data evolving from the various experiments outlined above.

8. PRINCIPAL CONTRIBUTORS

The principal contributors to this contract during the past quarter have been A. O. Jensen, D. Worden, H. Todd, A. E. Campbell, and W. Dong.

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APPENDIX A

CHEMICAL CLEANING SPECIFICATION

MATERIAL: TANTALUM OR MOLYBDENUM

MATERIALS USED:

Ethyl alcohol (CH_3CH_2OH)
Hydrochloric acid (HCl)
Chromic acid (saturated solution of potassium dichromate in hot concentrated sulphuric acid)
Distilled water

PROCEDURE

Step No. 1

Place part in beaker of ethyl alcohol. Place beaker in ultrasonic cleaner. Clean for approximately 60 seconds.

Step No. 2

Remove parts from ethyl alcohol - allow to dry.

Step No. 3

Place parts in beaker of hot hydrochloric acid for approximately 30 seconds.

Step No. 4

Remove from hydrochloric acid and rinse in beaker of boiling distilled water for approximately 2 minutes.

Step No. 5

Remove from distilled water - allow to dry.

Step No. 6

Dip in ethyl alcohol - remove and allow to dry.

Step No. 7

Place parts in beaker of hot chromic acid ($110^{\circ}C$).

Step No. 8

Remove from chromic acid and rinse vigorously in large beaker of cold distilled water.

Step No. 9

Rinse parts in hot distilled water for approximately 2 minutes.

Step No. 10

Rinse parts in cold distilled water for approximately 10 seconds.

Step No. 11

Remove parts from distilled water - allow to dry.

Step No. 12

Rinse parts in clean ethyl alcohol - remove and allow to dry.

Step No. 13

Wrap parts in lint free paper. Place in desiccator until ready for use.

CAUTION

All chemical cleaning will be done under the fume hood at all times. Caution should be taken in that the vapors are harmful to the skin and clothes. Rubber gloves and tongs should always be used. The tap water in the fume hood should be kept running constantly in case of any accident. Avoid the inhalation of any of the fumes.